Flight Experiment to Study Double-Diffusive Instabilities in Silver-Doped Lead Bromide Crystals

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<u>Objectives</u>

The main objective of the present program is to understand thermosolutal convection during crystal growth of PbBr₂-AgBr alloys. This involves identification of the growth conditions for microgravity experiments delineating the microsegregation, observation of convecto-diffusive instabilities, and comparison with theoretical models. The overall objectives can be summarized as follows:

- Observe and study the double diffusive and morphological instabilities in controlled conditions and compare with theoretically predicted convective and morphological instability curves.
- Study the three dimensional morphological instabilities and resulting cellular growth that occur near the onset of morphological instability in the bulk samples under purely diffusive conditions.
- Understand the micro- and macro-segregation of silver dopant in lead bromide crystals in microgravity.
- Provide basic data on convective behavior in alloy crystals grown by the commercially important Bridgman crystal growth process.

Necessity of Microgravity

Lead bromide doped with silver can be grown under normal gravity conditions, but the double diffusive nature of the convection will cause mixing of the molten charge material. This, in turn, will cause the solute composition in the crystal to constantly increase during growth. In semiconductor devices, where the electronic properties are a function of the crystal composition, this constant compositional variation is undesirable.

During the solidification of doped materials in Bridgman geometry, generation of destabilizing temperature gradients in the melt is unavoidable, resulting in buoyancy-induced convective mixing of the liquid phase. On earth this mixing is generally very intensive and prevention of convection is important in order to minimize micro- and macro-segregation and to obtain homogeneous properties throughout the solidified material. In an actual furnace it is extremely difficult to

eliminate the radial temperature gradient completely. Unavoidable gradients may give rise to flows, which lead to lateral segregation in the solidified material. In binary systems, if the solute pile up ahead of the solidification front is lighter than the solvent, this alone would cause positive density gradient. The net density gradient can have various profiles, depending on the properties of the melt (such as thermal conductivity) and diffusion coefficient or growth conditions (such as growth rate and thermal gradient). Even if the net resulting temperature gradient is stable, convection can occur due to double diffusive character of solute and temperature with different diffusivities. While there have been many observations on earth of this phenomena in thin samples where convection is not important, it is nearly impossible to study three-dimensional instabilities in bulk samples on earth under purely diffusive conditions. The space experiment on transparent lead bromide-silver bromide alloys would permit a study of the various three-dimensional morphologies that occur near the onset of morphological instability. Since the lead bromide-silver bromide system is transparent, experiments in space would allow the direct observation of morphological instability and the resulting cellular growth.

The present experiment on lead bromide-silver bromide alloys permits a study of various three-dimensional morphologies that occur near the onset of morphological instability in a bulk crystal grown by solidification technique. Being able to see exactly what is happening during growth in low earth orbit makes this a unique system for microgravity experimentation. The system chosen here has dual advantages: (a) lead bromide is a transparent system almost ideally suitable for direct *in situ* observations to study solid-liquid interface phenomena; and, (b) lead bromide holds great promise for technological applications of acousto-optic devices and narrow band ultraviolet filters.

Significant Results

The solidification experiments with PbBr₂-AgBr alloys (500 and 5000 ppm) showed doublediffusive instabilities at the solid-liquid interface. When the sample was held stationary, any convection present in the liquid was attributed to the radial heat losses. A systematic observations at the interface showed the development of the depression which finally ends in interfacial breakdown. When we repeated this experiment with pure lead bromide at a speeds of 2.5 cm/day, the interface remained flat and did not show any instabilities. The interface got depressed in the center and then slowly formed the instability. As a function of time, the instability developed with a much larger amplitude. When the translation velocity was increased, the interface started breaking down. The flow pattern observed in the PbBr₂-AgBr system can be described as a "toroidal roll." A slight asymmetry of the system resulted in the displacement of the node and axis of the tours from the central axis of the tube. When the toroidal flow persisted for many hours and the tube was moving, the interface was observed to be pinched where the radial inflow converged leading to the line defect. A theoretical calculation was performed to generate the concentration profile for the solute distribution and we are comparing it with the experimentally measured values. We measured the diffusion coefficient and thermal conductivities of solid and liquid which were used in computing stability curves and solute distribution in the crystal.